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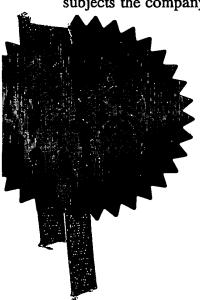
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## Patents Form 1/77

Patent 1977 (Rule 16)



19JUNGS EB1-239-1 D01091 P01/7700 0200-0314242.9

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AA 1623 GB

2. Patent application number (The Patent Office will fill in this part)

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0314242.9

3. Full name, address and postcode of the or of

each applicant (underline all surnames)

JOHNSON MATTHEY PUBLIC LIMITED COMPANY

2-4 COCKSPUR STREET TRAFALGAR SQUARE LONDON SW1 5BQ

Patents ADP number (if you know it)

If the applicant is a corporate body, give the GB country/state of its incorporation

536268009

3991411001

f. Title of the invention

FLUID TREATMENT

5. Name of your agent (if you have one)

ANDREW DOMINIC NUNN

"Address for service" in the United Kingdom to which all correspondence should be sent (tricluding the postcode)

JOHNSON MATTHEY TECHNOLOGY CENTRE

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Country

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Date of filing
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Number of earlier application

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# **FLUID TREATMENT**

This invention relates to fluid treatment by passage through a bed of material presenting contacting surfaces, in particular of material capable of effecting a chemical change in a constituent of the fluid.

In such treatment it is often found that efficiency decreases with time but can be regenerated by intermittent change of operating conditions and/or chemical composition. This may require interruption of the treatment and/or provision of a plurality of treatment reactors. The result is increased cost, increased space requirement and possible control problems. Such a result is especially undesirable in applications such as automotive exhaust treatment.

In the following description and claims: terms based on the word 'sorb' will be used to denote 'absorb' or 'adsorb' or any occurrence of both such processes; and metal compounds effective to sorb NOx will be referred as 'oxides', with the understanding that this term includes other oxidic compounds such as hydroxides and carbonates effective as NOx sorbents and present in the conditions of exhaust gas treatment. the term 'regenerand' will be used to denote a bed-region about to be subjected to a regeneration step.

According to the invention in a first aspect a process of fluid treatment by passage through a bed of material presenting contacting surfaces and subject to decrease of activity with time is characterised by using a bed subdivided in the direction of fluid flow into a plurality of mutually fluid-tight bed-regions and successively subjecting a sub-set of such regions to regeneration.

According to the invention in a second aspect a system for fluid treatment by passage through a bed of material presenting contacting surfaces and subject to decrease in activity with time comprises a reactor in which such bed is subdivided in the direction of fluid flow into a plurality of mutually fluid-tight bed-regions and means for successively subjecting a sub-set of such bed-regions to regeneration.

The total number of bed-regions is most simply 2, but more can be used, for example up to 8 or more. At a given time some of the bed-regions can be neither in use nor regenerating, for example to provide capacity in reserve. Further, at a given time, all the bed-regions can be in use, none regenerating, especially in the change-over from one of said sub-sets of bed-regions to another. If the total number of bed-regions is 2, the change-over from treatment to regeneration can be effected by simple valving, for example a flap valve in the treatment vessel upstream of the treatment bed. If the total number of bed-regions is greater, selection of the regenerand bed-region can be by a deflector moveable in alignment therewith. The valve or deflector may direct fluid at its full flow rate into the regenerand bed-region or, preferably, at a decreased rate, in volumes per unit cross-sectional area per unit time, e g 5-50%, compared with its full rate entering the reactor.

Regeneration typically includes introducing a regenerant fluid into the regenerand bed-region. Preferably this is by injection of such fluid at location aligned with the regenerand bed-region. If fluid flow to the regenerating bed-region is at less than the full rate as mentioned above, this limits the rate of flow from that bed-region and affords the advantage that, since such fluid commonly contains residual unreacted regenerant, less has to be emitted to atmosphere or treated further, e g in engine exhaust treament by feeding it to the engine fuel+air inlet by exhaust gas recycle EGR as described in our co-pending application internal ref AA 1622.

The bed is suitably a monolith or plurality of monoliths, for example foamed, moulded or extruded ceramicor wound-corrugated metal or metal as foam, sinter or ordered or random wire or flat wire. If the bed material permits fluid flow tranverse to the main direction of flow, the bed-regions should be separated by fluid-tight partitions.

General examples of the process and system are:

1. addition or removal of a component by respectively taking such component from, or sorbing it into, bed material;

2. catalytically effecting a chemical change when the fluid is not chemically at equilibrium *per se* or owing to added component; regeneration may be necessary to restore the activity of the catalyst;

# Specific examples of 1 are:

- a. NOx-removal from exhaust from a lean-burn reciprocating internal combustion engine by sorbing NOx in a basic material to produce nitroxy salt, then regenerating such material by intermittently contacting it with reductant or NOx-specific reactant. Such reductant is non-selective, for example hydrocarbon e.g. engine fuel or oxygenated hydrocarbon. Such NOx-specific reactant is selective, for example a nitrogen hydride e g ammonia or hydrazine, introduced as such or with water or as a precursor such as urea. The NOx sorbent is typically selected from:
- (i) oxides of alkali-, alkaline earth-, rare earth- and transition-, metals capable of forming nitroxy salts of adequate stability in sorbing conditions and of releasing /reacting nitrogen oxides in regenerating conditions.
- (ii) adsorptive materials such as zeolites, carbons and high-area oxides. Whichever compounds are used, there is preferably present one or more catalytic agents such as precious metals, especially Pt+Rh, effective to promote reaction of NOx with reductant or NOx-specific reactant.
  - b. SOx-removal analogously;
- c. collecting particulate matter from such exhaust by impingement or filtration, then regenerating by combusting the collected PM. The temperature at or above which such combustion takes place is known as the 'balance temperature. Combustion may be assisted by introduction of NO<sub>2</sub> or ozone or plasma or readily combustible liquid;
- d. contacting the fluid with adsorbent such as zeolite, to remove a component or to add a component; regeneration is respectively by expelling adsorbed component from, or recharging, the bed material.

Specific examples of 2 are exhaust treatment by one or more of 3-way catalysis, oxidation of HC/CO, oxidation of NO to NO<sub>2</sub>.

Since the process during regenerations is operated with less than the whole of the volume of the contacting beds, the volume of each bed and the timing of the start and duration of regeneration should be matched so that the treatment capacity is not exceeded.

The invention is illustrated by the accompanying drawings, in which:

Figure 1 is a schematic sectional view of a NOx sorber embodying the invention, in combination with a soot combustion reactor, as applicable to the exhaust of a diesel engine;

Figure 2 shows a NOx sorber similar to that of figure 1 but using a 2-part soot combustion reactor, the second part housing a flow-reversing filter.

Referring to Figure 1, item 120 is a soot combustion reactor the inlet of which is connected to the exhaust manifold of a diesel engine (not shown). Reactor 120 at its upstream portion contains oxidation catalyst 122 consisting of a ceramic honeycomb carrying a washcoat and Pt. At its downstream portion reactor 120 contains PM filter the passages of which are 124, consisting of filter-grade ceramic honeycomb alternatingly open and closed at the inlet end and, corresponding to the inlet open passages, alternatingly closed at the outlet end. From the outlet end of reactor 120 plenum 126 continues as the operating chamber of flap valve 128X,Y, Z at the inlet of NOx sorber vessel 130. Vessel 130 contains NOx sorber 131X,Y consisting of a throughflow ceramic honeycomb carrying an alumina washcoat containing barium oxide and metallic Pt+Rh. The fulcrum of flap valve 128X,Y,Z is mounted on partition 129 which extends diametrally across the face of reactor 130 and is gas-tightly sealed to the face of sorber 131. Each region X,Y of reactor 130 either side of valve 128 is provided with reactant injector 132X,Y. In the complete reactor 130 as shown valve 128 is in the central position C. Valve positions X and Y are shown as insets.. Reactor 130 is formed with outlet 134, leading to atmosphere or to further treatment.

In the normal operation of the system, the exhaust gas, comprising steam,  $N_2$ ,  $O_2$ ,  $CO_2$ , HC, CO, NOx and PM, at e g 300C contacts catalyst 122 over which NO is oxidised to  $NO_2$  and some of the HC and CO are oxidised to steam and  $CO_2$ . It then

enters filter 124 on which most of the PM is collected and combusted by reaction with the NO<sub>2</sub> formed in catalyst 122 and possibly with O<sub>2</sub>. The PM-freed gas then undergoes treatment in one of the 3 modes: 128Z: sorber regions 130X and 130Y both sorb NOx; 128X: region 131X receives a small fraction of the gas leaving plenum 126 and injection of HC at 132X. It undergoes regeneration; and its effluent is reunited with that of region 130Y; region 131Y receives the major portion of the gas, sorbs NOx and passes its effluent to atmosphere at 134; 128Y: region 131Y performs the duty described at 128X.

The engine management system (not shown) changes from region X to region Y when sorber 131Y has NOx sorption capacity in hand; and vice versa.

Referring to Figure 2, reactor 120 now only contains catalyst 122. The PM filter vessel, now numbered 125, is in separate vessel 121. It differs in providing for reversal of the direction of flow through the filter. The outlet of reactor 120 is connected to filter vessel 121 by way of 4-way valve 123, operable in positions 123A and 123B (inset), to give respectively RH to LH and LH to RH flow through filter 125, but with no blocking mid-point; since the midpoint bypasses filter 125. Operation of valve 123 is controlled to be very rapid. Leaving filter 125 in either direction, the gas passes through valve 123AorB to NOx sorber 130, which is structurally and functionally as in figure 1.

## **CLAIMS**

- 1. A process of fluid treatment by passage through a bed of material presenting contacting surfaces and subject to decrease of activity with time, characterised by using a bed subdivided in the direction of fluid flow into a plurality of mutually fluid-tight bed-regions and successively subjecting a sub-set of such regions to regeneration.
- 2. A system for fluid treatment by passage through a bed of material presenting contacting surfaces and subject to decrease in activity with time comprising: a reactor in which such bed is subdivided in the direction of fluid flow into a plurality of mutually fluid-tight bed-regions; and means for successively subjecting a sub-set of such bed-regions to regeneration.
- 3. A process according to claim 1 or a system according to claim 2 operable additionally in modes in which some of the bed-regions are neither in use nor regenerating, or in which all the bed-regions are in use, none regenerating.
- 4. A process or system according, as appropriate, to any one of the preceding claims, in which the total number of bed-regions is 2 and the change-over from treatment to regeneration is effected by a flap valve in the treatment vessel. upstream of the treatment bed.
- 5. A process or system according, as appropriate, to any one of claims 1 to 3, in which the total number of bed-regions is greater than 2 and selection of the regenerand bed-region is by a deflector moveable in alignment therewith.
- 6. A process or system according to any one of the preceding claims in which fluid directed into the regenerand bed-region flows at a rate lower than that of the fluid entering the vessel.

- 7. A process or system according to any one of the preceding claims in which regeneration includes introducing a regenerant into the regenerand bed-region.
- 8. A process or system according to claim 7 in which regenerant introduction is by injection of regenerant fluid at location(s) aligned with the regenerand bed-region(s).
- 9. A process or system according to any one of the preceding claims in which the bed is a monolith or plurality of monoliths comprising foamed, moulded or extruded ceramic, wound-corrugated metal or metal as foam, sinter or ordered or random wire or flat wire, and including, if the bed material permits fluid flow tranverse to the main direction of flow, separation of bed-regions by fluid-tight partitions.
- 10. Use of the process or system according to any one of the preceding claims for addition or removal of a component by respectively taking such component from, or sorbing it into, bed material; or catalytically effecting a chemical change.
- 11. Use according to claim 10 in any of:
- a). treating the exhaust of a lean-burn reciprocating internal combustion engine by sorbing NOx in a basic oxidic material to produce nitroxy salt, then regenerating such material by intermittently contacting it with reductant or NOx-specific reactant;
- b) treating the exhaust of a lean-burn reciprocating internal combustion engine by sorbing SOx in a basic oxidic material to produce sulfoxy salt, then regenerating such material by intermittently contacting it with reductant;
- c). treating the exhaust of a lean-burn reciprocating internal combustion engine by collecting particulate matter from such exhaust by impingement or filtration, then regenerating by combusting the collected PM by one or more of: increasing temperature to the 'balance temperature'; injecting NO<sub>2</sub> or ozone or plasma; injecting readily combustible liquid;
- d). contacting the fluid with adsorbent to remove a component or to add a component, then regenerating respectively by expelling adsorbed component from, or recharging, the bed material.

12. Use according to claim 10 in engine exhaust treatment by one or more of 3-way catalysis, oxidation of HC/CO, oxidation of NO to NO<sub>2</sub>.

# **FLUID TREATMENT**

# Abstract

A process for fluid treatment by passage through a bed (130) of material presenting contacting surfaces and subject to decrease of activity with time, characterised by using a bed subdivided in the direction of fluid flow into a plurality of mutually fluid-tight bed regions, (131X; 131Y) and successively subjecting a sub-set of such regions to regeneration.

[Figure 1]

